Femtosecond Synchrotron Pulses from the Advanced Light Source

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An important new area of scientific research in chemistry, physics, and biology is the investigation of ultrafast structural dynamics in condensed matter using femtosecond x-ray pulses. The dynamic properties of materials are governed by atomic motion which occurs on the fundamental time scale of a vibrational period, ~100 fs. This is the time scale of interest for ultrafast chemical reactions, non-equilibrium phase transitions, vibrational energy transfer, surface desorption and reconstruction, and coherent phonon dynamics. To date, our understanding of these processes has been limited by lack of appropriate tools for probing the atomic structure on an ultrafast time scale. X-ray techniques such as diffraction and EXAFS yield detailed information about "static" atomic structure. However, the time resolution of high-brightness synchrotron x-ray sources such as the Advanced Light Source (~30 ps) is nearly three orders of magnitude too slow to directly observe fundamental atomic motion. Conversely, femtosecond lasers measure transient changes in optical properties of materials on a 10 fs time scale, but optical properties are only indirect indicators of atomic structure.

We have recently demonstrated a novel scheme for generating ultrashort pulses of synchrotron radiation[1]. Our approach is to create femtosecond time-structure on a long electron bunch by using a femtosecond laser pulse to modulate the energy of an ultrashort slice of the bunch. The modulation is achieved via interaction between the electrons and the light field in a wiggler (Figure 1a). Half of the electrons overlapping with the ultrashort pulse are accelerated and half are decelerated depending on the optical phase. The laser-induced energy modulation is several times larger than the rms beam energy spread at the ALS (~1.8 MeV), and only an ultrashort slice of the electron bunch experiences this modulation. The accelerated and decelerated electrons are then spatially separated from the rest of the electron bunch (in a dispersive bend of the storage ring) by a transverse distance that is several times larger than the rms transverse size of the electron beam (Figure 1b). Finally, by imaging the displaced beam slice to the experimental area, and by placing an aperture radially offset from the focus of the beam core, we will be able to separate out the radiation from the offset electrons (Figure 1c).

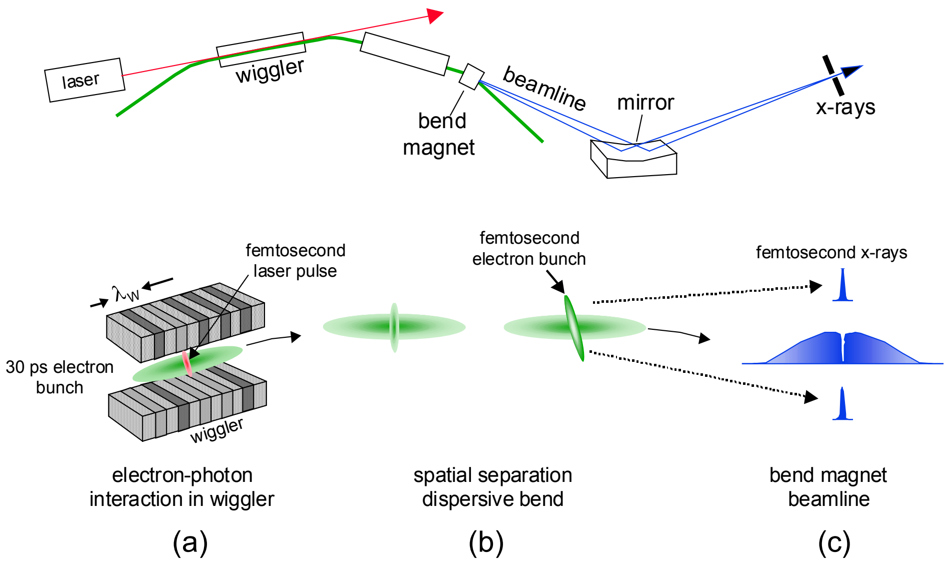

Figure 1. Schematic illustration of the technique for generating femtosecond synchrotron pulses. (a) laser/electron beam interaction in resonantly-tuned wiggler, (b) separation of accelerated femtosecond electron slice in a dispersive (bend) section, (c) generation of femtosecond x-rays at a band-magnet beamline.

This technique has recently been implemented on ALS beamline 5.3.1. A cryogenically cooled Ti:sapphire femtosecond laser system is located near the beamline endstation, and the laser pulses (~1 mJ, 100 fs pulse duration, 2 kHz repetition rate) are projected across the storage ring roof blocks to sector 5, where they enter the main vacuum chamber through a back-tangent optical port. Amplified femtosecond pulses co-propagate with the electron beam through wiggler W16 in sector 5. A photon stop/mirror following the wiggler reflects the laser light and the visible wiggler emission out of the vacuum chamber for diagnostic purposes in order to verify the spatial, spectral, and temporal overlap of the laser beam with the fundamental (undulator-like) emission from the wiggler.

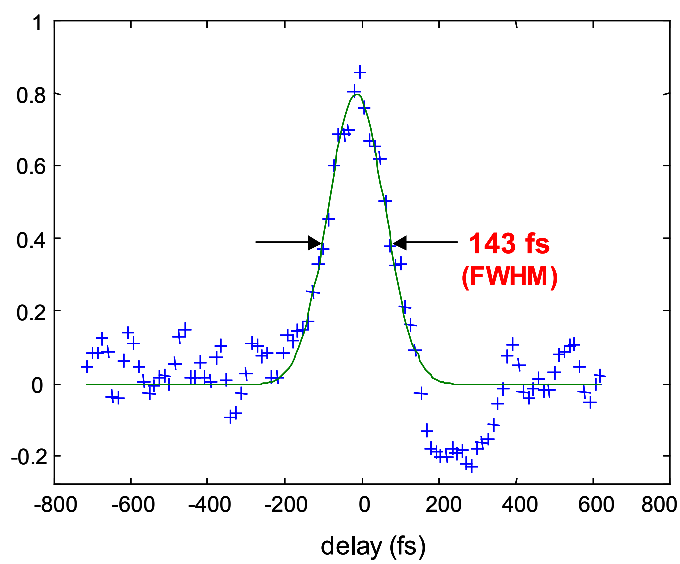

Figure 2. Cross correlation of an ultrashort synchrotron pulse with femtosecond laser pulse. The deconcolved synchrotron pulse duration is 143 fs (FWHM).

Femtosecond duration synchrotron pulses are directly measured by cross-correlating the visible light from bend-magnet beamline 5.3.1 at the ALS with a delayed laser pulse in the nonlinear crystal, BBO. Figure 2 shows a laser synchrotron cross-correlation measurement corresponding to synchrotron pulses of <150 fs duration, the shortest pulses ever generated from a synchrotron. Additionally, these measurements were made during normal multi-bunch operation of the storage ring, which illustrates the compatibility of this approach with the requirements of other synchrotron users, notably the protein crystallography beamline which simultaneously uses the wiggler W16. An important point is that the femtosecond time structure will be invariant over the entire spectral range of bend-magnet emission from the near infrared to the x-ray regime, making this a very powerful tool for femtosecond spectroscopy.

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